Occurrence and Significance of Pesticide Residues in Water[1]

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Man throughout the civilized world is rapidly coming to realize that environmental contamination, with its harmful ecological implications, is a matter to be taken seriously. I bring to your attention one facet of environmental contamination; namely, water pollution by pesticides. It is but another example of the adage that a good thing in the wrong place can be undesirable.

The Problem

Water pollution by pesticides became a problem in the 1940's concurrently with rapid advances in pest control made possible by the development of new synthetic toxicants. Many of these synthetics are remarkably lethal to aquatic forms of life. Farmers were startled at the sudden loss of fish in their ponds and streams following rains sufficient to cause runoff from treated cropland (Young and Nicholson, 1951). Aerial applications of DDT to control forest insects were quickly followed by losses of valuable sports fish and the aquatic insects upon which they fed (Hoffman and Drooz, 1953; George, 1959).

Today we still experience periodic losses, but we know considerably more than formerly about their causes and prevention. We know that sublethal quantities of pesticides, primarily chlorinated hydrocarbon insecticides, occur widely and frequently in our streams, lakes, and even in the sea. This occurrence is indirectly evident through the recovery of residues from the tissues of fish (Nicholson, 1967; Anon., 1963), and directly evident by chemical analysis of water. In an effort to determine the extent of pesticide pollution throughout the United States, Weaver *et al.* (1965) examined water samples in September 1964 from 56 rivers and 3 of the Great Lakes. Chlorinated hydrocarbon in-insecticides were found in 44 rivers and in Lake Michigan at Milwaukee at concentrations ranging from 0.002 to more than 0.118 μg/liter. Dieldrin was found in 39 rivers and Lake Michigan; DDT, or its metabolite DDE, was found in 25 rivers; and endrin was found in 22 rivers and Lake Michigan.

The two principal sources of water contamination by pesticides today are runoff from the land and discharges of industrial wastes. Other causes are (a) activities intended to control aquatic life (plants, fish, or insects), (b) carelessness and accidents.

Runoff from the Land

Consider first insecticide runoff from the land. In 1959 my laboratory undertook to follow the course of water pollution by insecticides in a single large agricultural watershed over a period of nearly seven years (Nicholson *et al.*, 1966). We selected a 400-square-mile cotton-growing area in northern Alabama in which cotton acreage varied annually from 13,000 to 16,000 acres. From 8 to 84% of this acreage was treated with insecticides each summer depending upon the degree of boll weevil and

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bollworm infestation. The quantity of technical grade insecticides used each year varied from 12,000 to 140,000 pounds. Toxaphene, DDT, and BHC accounted for 84–99% of all usage.

Water sampling was done nearly continuously at a municipal water treatment plant situated at the downstream end of the river basin. Thus, water samples represented drainage from the entire study area. We learned the following:

(a) Insecticides did run off the land. They entered the river from the watershed in general, rather than from a few favorably located cotton fields.

(b) Toxaphene, DDT and BHC were recovered in water samples in concentrations generally less than 1 μg/l. Highest mean recoveries were usually made during the summer, the season of application.

(c) Nearly all water samples contained insecticides year around during years of heaviest application. Toward the end of two years of minimal application (12,000 and 14,000 lbs., respectively), the frequency of negative water samples increased, indicating an improvement in river water quality with diminished insecticide usage.

(d) Toxaphene and BHC, first and third in poundage applied, were the most frequently found in water. DDT, which constituted 26–35% of the pesticides used, was recovered only during the fifth and sixth years of observation.

(e) DDT exhibited a marked affinity for sediment, and suspended sediment was the primary vehicle for its transport, thus accounting for its tendency to appear less frequently in water. Toxaphene and BHC, in contrast, were found much less frequently in association with sediment and were transported primarily in solution in the water.

A study by Bailey and Hannum (1967) reported from California sheds further light on runoff as a means of pesticide transport. Approximately 20% of all pesticides used in the United States annually is applied in California. The areas studied included the agriculturally important Imperial, San Joaquin, and Sacramento Valleys, where irrigation is required for successful farming.

Major findings were:

(a) DDT, DDD, toxaphene, heptachlor epoxide, lindane, dieldrin, and BHC were found both in surface water and in tile drainage water in concentrations generally less than 1 μg/l.

(b) All aforementioned insecticides, except BHC, were found in sediment and ranged from 1 to 1200 μg/l.

(c) Thiophosphate insecticides, which degrade more readily, were detected primarily in agricultural drainage, irrigation wastewater, and surface water directly associated with insecticide applications.

(d) Pesticide concentrations were highest in agriculturally developed areas and decrease in surface water in proportion to inflow dilution and uptake by sediment and aquatic organisms.

Manufacturing Wastes

Manufacturing wastes also may contain quantities of pesticides sufficient to have a decided impact on water quality. The types of industries involved include producers of basic pesticides, cooperage firms that reclaim used pesticide drums, and textile plants that mothproof woolen yarns and fabrics with dieldrin. These plants usually have liquid wastes requiring disposal—wastes which frequently contain residues of unrecovered pesticides.

Virtually all of these industrial plants provide some sort of waste treatment, but it is not always as effective as it should be. Dilution in the receiving stream cannot be depended upon to eliminate the impact of the waste load. Sublethal residues of chlorinated hydrocarbon insecticides can undergo a buildup in biotic components of the receiving water body, and ordinarily sublethal quantities of organophosphate pesticides may, through extended exposure, progressively inhibit the acetylcholinesterase enzyme to a degree that can kill aquatic life. Direct and catastrophic damage also has occurred when in-plant trouble resulted in an unanticipated slug discharge of wastes containing concentrations of a pesticide sufficient to be acutely toxic. An example of such a situation and its successful management follows.

A plant in Alabama which manufactures parathion and methyl parathion experienced a breakdown in its waste treatment facility in May 1961 (Anon., 1961). Process wastes were discharged to the sewerage system of an adjacent city and approximately 60% of the combined sewage and industrial waste was diverted, untreated, to a small stream during the breakdown. Fish, turtles, and snakes died along 28 miles of the stream, the average discharge of which at the time was 211 million gallons a day at a velocity of three-fourths mile per hour. The creek entered the Coosa River the average discharge of which was then about 28 times greater than that of the creek. Yet even with that dilution, parathion residues were recovered 90 miles down the Coosa and some lesser fish kills occurred in it. After a second fish kill in 1966, the company constructed a basin for temporary containment of its wastes, should another emergency arise. This simple device, along with the usually adequate waste treatment normally provided, should effectively prevent recurrence of the problems previously experienced.

Accidents and Carelessness

Perhaps the third most significant cause of pesticide pollution lies in accidents and accident's handmaiden, carelessness. Intensive educational campaigns sponsored by agricultural, conservation, water-pollution-control, and public health agencies and by the agricultural chemicals manufacturing industry have reduced the frequency of such occurrences. Most farmers have learned that it is inadvisable to dump unused spray residue where it might run into a waterway, and that they should not wash out spray equipment in a creek. Aerial applicators now pay heed to the protection of ponds and rivers. Nevertheless, some instances of water pollution by pesticides still occur as a result of thoughtlessness and accidents. An instance in which human health was at stake will serve as an example (Anon., 1964).

In 1964 a rancher instructed his hired hand to dispose of approximately fifty 4-lb. bags of over-age 15% parathion dust. Unknown to the rancher, this was done by dumping the bags off a highway bridge into the Peace River one mile upstream from the municipal water intake of Arcadia, Florida, a town of about 6,000 people. The act was discovered when some boys fishing near the bridge hooked a bag and had the foresight to report it.

The town fortunately had an auxiliary well for emergency use and immediately reverted to it. The citizens were instructed not to use the water, and flushing of the mains was begun. Subsequent analysis of water samples showed that the parathion concentration in the distribution system after flushing was generally less than 1 μg/l. However, a series of samples taken from a tap at the local bus station contained amounts up to 380 μg/l.

Investigation revealed that the bags of parathion had been dumped in the river about 10 days before their discovery. The bags were polyethylene lined and resisted rapid disintegration. Many were recovered unbroken and those that did disintegrate apparently did so intermittently over a period of several weeks. This may have been the reason that residue levels sufficiently high to be a threat to human health or the fish in the river did not occur. All but 8–12 bags were eventually found. Parathion residue occurred in river water for about two weeks after discovery at concentrations generally less than 1 μg/l.

Control of Aquatic Life

The chemical control of aquatic weeds, rough fish, and aquatic insect pests is generally managed by professionals so that undesirable consequences are minimized. A need currently exists for herbicides approved for broader use in water. A joint committee of the Departments of the Interior and Agriculture are seeking a solution to this need. A similar, but remotely related, source of pesticide residues is poaching for fish. We still have instances where insecticides, frequently toxaphene or DDT, are illegally released in water to catch fish.

Ground Water Pollution

No broad discussion of this subject would be complete without considering ground water. The potential for pesticide contamination of ground water is very much less than for surface water. However, it can occur.

A case is on record in Florida where the municipal water supply wells of a city of 25,000 contained low levels of parathion (usually less than 1 μg/l) over a several month period in 1962-63. The city's water supply consisted of both surface water, which reached the municipal water treatment plant via a canal from a citrus fruit producing area, and of five wells which were located in the vicinity of the treatment plant. The water from both sources contained parathion. The wells were rather shallow—drilled to a depth of about 100 feet and screened both at the bottom and at about the 30- to 50-foot levels. It is speculated that heavy pumping from the wells drew down surface water from the canal.

A more serious instance occurred in the South Platte River Basin near Denver, Colorado in the mid-1950's, caused by seepage of 2,4-D and related compounds from an industrial waste lagoon (Cottam, 1960). Water from wells in a 6.5-square-mile area when used for irrigation was sufficiently contaminated to cause crop damage.

Eye (1968) concluded after a study of the physical-chemical behavior of dieldrin in the soil that residues of this insecticide cannot be transported in significant amounts through soils into subsurface water by infiltration, and therefore they pose no threat to the quality of ground water. We have examined many well water samples from the Southeastern States and only in a few instances have we detected any evidence of chlorinated hydrocarbon insecticides. In those few cases, I recall only two in which direct contamination did not seem to be a possible cause. On the other hand, Bailey and Hannum (1967) in California reported recovering a broad range of chlorinated hydrocarbon insecticides. In those few cases I recall underground tile drains from irrigated cropland. They did not speculate on how insecticides entered the drains. A possible route might be through cracks or other direct passages from the surface.

In several of our mid-western States, where water of high quality is in limited supply, consideration is being given to using runoff water collected seasonally in playa lakes as a source from which to recharge ground water aquifers. The Robert S. Kerr Water Research Center of the Federal Water Pollution Control Administration at Ada, Oklahoma, is engaged in studies to determine the quality of recharged water, including the persistence and distribution of pesticides that may be contained in such water, after being pumped into the ground for storage.

Significance

We have seen that water contamination by pesticides occurs widely and commonly at concentrations generally less than 1 μg/l. Higher concentrations occur intermittently. But of what significance are such occurrences?

Aquatic Life

Quite clearly the unintentional killing of fish and other aquatic life by overwhelmingly lethal concentrations of pesticides is harmful and undesirable. Occurrences of this type are generally local, readily apparent, and sporadic, with partial or total repopulation quickly occurring.

Widespread, long-term, low-level contamination of the environment is much more difficult to evaluate and is a matter of growing public concern. It is caused primarily by a few compounds, members of the chlorinated hydrocarbon insecticide group—the so-called "hard" insecticides—that persist so long in nature and therefore escape our control after they are applied. Other pesticides, by and large, either degrade with reasonable rapidity or are so restricted in usage as to be of less concern except in special cases. One is tempted to speculate as to whether we would have had the public outcry over pesticides that we have experienced during the past 10 to 15 years had it not been for these few "hard" insecticides. I am inclined to think that it would have been much less extensive.

The single sublethal manifestation with chlorinated hydrocarbons that is most obvious, and the significance of which is least understood, is that of biological accumulation. Biological accumulation may occur through direct absorption from the water or by absorption and passage through the food chain. The implications for damage are great, but well defined examples of proved harm are few, perhaps because biological accumulation is not as generally damaging as feared, but also perhaps because the ecological relationships involved are so extremely complex that they are difficult to unravel.

Light has been cast on this phenomenon by numerous researchers. Cope (1965), investigating the distribution of DDT through various compartments of a simplified ecosystem, reported that two weeks after the application of ^{14}C–DDT at a concentration of 20 μg/l to aquarium water, the water contained 0.42 μg/l, soil contained 6 μg/kg, and vegetation contained 15,600 μg/kg. Two weeks after fish were placed into the aquaria, they contained 1,000 μg/kg of DDT. Woodwell *et al.* (1967) investigated biological concentration of DDT among various trophic levels of a Long Island salt marsh and reported values increasing from 0.04 mg/kg in plankton to 75 mg/kg in ring-billed gulls. Highest concentrations occurred in scavenging and carnivorous fish and birds, although the birds had 10–100 times more than the fish. Gakstatter and Weiss (1967) exposed bluegills and goldfish in aquaria to ^{14}C–DDT, dieldrin, and lindane to study uptake, retention, and release by the fish. They showed that the lindane was entirely released within two days and that more than 90% of the initial dieldrin was eliminated within two weeks. However, more than 50% of the DDT was still retained after 32 days. More significant, they showed that DDT and dieldrin were readily transferred from contaminated to uncontaminated fish held in clean water. Apparently, some of the persistent insecticides are capable not only of undergoing biological magnification but also of cycling between water and the organisms living in it.

The accumulation of pesticides in the bodies of fish has been cited as the probable cause of the secondary poisoning of a variety of fish-eating birds. The most notable example is that described by Hunt and Bischoff (1960) in which western grebes overwintering on Clear Lake, California died, presumably from eating fish containing high DDD residues. Keith (1966) reported an unusually high mortality of fish-eating birds between 1960 and 1962 at the Tule Lake National Wildlife Refuge in California, which he attributed, circumstantially, to ingestion of toxaphene accumulated in fish. A study of this refuge in 1965–66 (Godsil and Johnson, 1968), when endrin was the principal insecticide used on the nearby irrigated farmland, indicated a marked increase of endrin in all trophic levels during the crop-growing season (May-Sept.) with a subsequent decline to near or below detectable limits in the off season. Fish accumulated maxima of 97 μg/kg in 1965 and 107 μg/kg in 1966. Endrin was not established as a permanent residue, and no wildlife losses were recorded. It is apparent that the so-called "hard" insecticides are not equally accumulative or persistent in food chain compartments.

Butler (1966 a, b, c) has done extensive work on the effects of low levels of chlorinated hydrocarbon insecticides on organisms of the marine environment. He showed that DDT in the water at levels as low as 1 μg/l caused a 20% reduction in oyster growth, and that oysters are efficient concentrators of DDT in their tissues. He believes that pesticides may be the cause of ill-defined but significant mortality, loss of production, and perhaps changes in the direction of natural selection in estuarine fauna. Cope (1965) concluded that exposure to sublethal amounts of DDT increases fish mortality by reducing resistance to other stresses.

Burdick and his co-workers (1964) in New York demonstrated that lethal amounts of DDT can be transmitted from female lake trout to their offspring through the egg. Lethality bore no relation to the concentration of DDT in the female. Fry died when the final contents of the yolk sacs were absorbed. These deaths occurred when the eggs contained DDT equivalent to 2.9 mg/kg or more of fry. This situation came to light when complete loss of lake trout fry occurred in 1955 and 1956 at a Lake George fish hatchery. It is a most subtle adverse effect that would be detected only under hatchery or laboratory conditions.

The influence upon the survival of aquatic organisms of transovarially conveyed pesticide residues is a subject worthy of further research. The period of dependence upon food stored in the egg sac may be for numerous fish species the most vulnerable period in their life histories as far as pesticides are concerned. If this is true, the chances are very slight that population losses would be directly observed in nature short of virtual elimination of a major species.

Much has been written about the effects of long-term exposure of aquatic organisms to pesticides at sublethal levels, but we still have a remarkably small amount of compellingly positive information indicating danger from organic chlorinated insecticides. DDT has received by far the most attention, possibly because its residues are so universally distributed. We need more research on other persistent insecticides. Although we do not have agreement within the scientific community concerning the danger of persistent residues in living organisms and in the environment, perhaps all can agree that it would be better if we did not have these uncontrolled residues.

Other Water Uses

In April 1968 the National Technical Advisory Committee on Water Quality Criteria of the Federal Water Pollution Control Administration submitted its first report to the Secretary of the Interior (Anon., 1968). This volume constitutes the most comprehensive document to date on water quality requirements for various uses. It contains recommendations for permissible limits for some pesticides.

The Subcommittee on Public Water Supplies based its recommendations on pesticides upon recommendations submitted by the Public Health Service Advisory Committee on Use of the Public Health Service Drinking Water Standards. The values were derived for that committee by an expert group of toxicologists and were established at those levels which, if ingested over extensive periods, could not cause harmful or adverse physiological changes in man. In the case of aldrin, heptachlor, chlordane, and parathion the values were set even lower than those physiologically safe, to avoid levels that could be tasted or smelled. Table 1 contains these recommendations.

Table 1. Surface Water Criteria for Pesticides in Public Water supplies (mg/l).[1]

	Permissible criteria	Desirable criteria
Aldrin	0.017	Absent
Chlordane	0.003	"
DDT	0.042	"
Dieldrin	0.017	"
Endrin	0.001	"
Heptachlor	0.018	"
Heptachlor epoxide	0.018	"
Lindane	0.056	"
Methoxychlor	0.035	"
Organic phosphates plus carbamates	0.1[2]	"
Toxaphene	0.005	"
2,4–D plus 2,4,5–T, plus 2,4,5–TP	0.1	"

[1] Adapted from Water Quality Criteria, Report of the National Technical Advisory Committee to the Secretary of the Interior, April 1968. Washington, D. C.

[2] As parathion in cholinesterase inhibition. It may be necessary to resort to even lower concentrations for some compounds or mixtures.

The subcommittees concerned with criteria for aquatic and wildlife (both freshwater and marine) and for agriculture each considered pesticides. The criteria, or formulae for determining criteria values, are generally too complex to justify discussion here, and the reader is referred to the original source.

An alternative suggestion for a water-quality criterion for fish, based on a group effect of about 100 organophosphorus and carbamate compounds, was derived at the Southeast Water Laboratory (Nicholson, 1967). This practical suggestion was based upon the ability of these compounds to inhibit acetyl-cholinesterase activity in the brains of fish. The degree of inhibition is a function of the compound, its concentration in water, and the duration of exposure. Death results from inhibition ranging from 40 to 70%. As little as 10% inhibition can be measured and statistically confirmed in a group of ten fish of the same species and of similar size. Therefore, it was suggested that 10% acetylcholinesterase inhibition in fish brain would serve as a good criterion of water quality involving chemicals capable of causing this inhibition. Unfortunately, no group effect for organochlorine insecticides has yet been developed upon which a similar criterion can be established.

Pesticide Pollution Control

The Southeast Water Laboratory has national responsibility within the Federal Water Pollution Control Administration for research leading to the control of pesticide pollution. Control is generally easiest at point sources; i.e., at industrial sources where waste effluent is discharged to a stream at a single outfall. We are currently beginning an inventory of waste-treatment practices at pesticide manufacturing and pesticide using industrial plants to establish a mutually beneficial relationship with some of these industries. Control may be accomplished by a variety of waste-treatment processes and by in-plant process changes. Effective control may be as simple as the provision of facilities for biochemical oxidation of effluents with auxiliary provision of a basin for containing extraordinary peak loads of wastes for more leisurely disposal. Our Laboratory is equipped with a variety of advanced analytical instruments, including a 100-megacycle high-resolution nuclear magnetic resonance spectrometer and a computerized mass spectrometer, with which we are able to determine the chemical nature of industrial waste effluents and assist in optimizing the design of advanced waste treatment systems.

The control of pesticide pollution associated with rural runoff is much more difficult to accomplish because its entrance into watercourses is not localized. Therefore, control must be accomplished by other means and ultimately rests in the hands of the users. Land-management practices designed to retard water runoff and soil erosion certainly are helpful measures. The retention of an untreated buffer strip adjacent to mountain streams was shown to prevent the runoff of DDT applied for forest insect control (Grzenda *et al.*, 1964).

We are conducting research with pure clay mineral model soils to develop basic concepts relative to the retention of representative pesticides on the land or their failure to be retained. Recently our scientists, cooperating with associates at Purdue University, demonstrated that s-triazine herbicides may be irreversibly adsorbed onto montmorillonite clay, and in so doing, undergo a chemical change to an innocuous compound (Russell *et al.*, 1968). Basic concepts developed are later confirmed with natural soils. The results frequently are directly applicable to rural runoff control recommendations.

Pesticide runoff from the land is directly related to runoff losses of both water and surface soil; the latter serve to transport pesticides from farm or forest to watercourses. Controlling this process are climatic, edaphic, hydrologic, physiographic, and cultural factors. If we knew more about the interplay of soil type, slope of the land, rainfall, and other climatic factors, cropping practices, and the behavior of the pesticides in use, we should be able to recommend measures to reduce the importance of rural runoff as a source of water pollution by pesticides. These recommendations might simply concern which pesticides to use or not to use in a given combination of circumstances. It might develop into water-pollution-control recommendations for geographic zones.

A comparable development has already been made in agriculture. I refer to the universal soil-loss equation that is applicable to guiding conservation farm planning throughout the United States (Wischmeier *et al.*, 1958; Wischmeier, 1969; Wischmeier and Smith, 1960, 1965). The factors upon which this equation is based are rainfall, soil-erodibility, slope length and gradient, cropping management, and erosion control practices. The possibility of extending the universal soil-loss equation and applying it to the prediction and control of pesticide pollution associated with rural runoff seems good and is being explored.

In the meantime, socio-economic developments are occurring outside the field of water pollution control that tend toward reduction of the water pollutional impact of the persistent organochlorine insecticides. The development of resistance to insecticides among cotton, corn, and sugarcane pests, to name a few, has forced total or partial abandonment of the formerly preferred "hard" insecticides in favor of more effective and, incidentally, less persistent types. Food and Drug Administration-controlled tolerance levels have required other changes. There is a growing public interest in environmental contamination control that may bring forth legislation outlawing the use of the "hard" insecticides as "hard" detergents were outlawed a few years ago. I should not like to see this happen, but would prefer to see substitutes used whenever it is feasible to do so, retaining the troublesome insecticides for use where they are absolutely necessary and where their usage will not result in further environmental contamination.

It is the responsibility of entomologists and leaders in the field of pesticide usage to take note, to look beyond the immediate problem of controlling insects, and to assume greater responsibility for preventing undesirable side effects resulting from the use of pesticides.

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